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Application of silica (SiO_2) nanofluid and Gemini surfactants to improve the viscous behavior and surface tension of water-based drilling fluids

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A B S T R A C T

Further studies into drilling fluids especially to reduce the use of oil and synthetic-based drilling fluids are ever-growing due to their contributions to environmental pollution. This study, therefore, attempts to evaluate the thermal, viscosity, surface tension, and filtration loss properties of water-based drilling fluids (WBDFs) upon the addition of Gemini surfactant-silica nanofluid. This surfactant-nanofluid was formed by dissolving silica nanofluid in the surfactant solution, and ultra-sonication was used to attain homogeneity. Characterization of the Gemini surfactant-silica (SiO_2) nanofluid was done by Fourier Transform Infrared Spectroscopy (FTIR). The viscosity, surface tension, and filtration loss properties were studied using the rheometer, tensiometer, and low-pressure, low-temperature (LPLT) filter press respectively. The experimental results showed that Gemini surfactants contributed to the highest increase in drilling fluid viscosity compared to a conventional surfactant. Also, when combined with silica-nanoparticles showed better thermal stability with an 11% average change in viscosity with increasing temperature and a decrease in surface tension and filtration loss both showing a 17% and 12% decrease respectively.

1. Introduction

A key determinant in the success of drilling operations is the formulation of highly functional drilling fluids. Drilling fluids carry out several purposes including removal of drilled cuttings and their transportation from the bottom of the hole to the surface, controlling the reservoir pressures, maintaining wellbore stability, and sealing off permeable zones in the formation with mud cake to prevent fluid loss [1]. According to World Oil's fluids nomenclature [2], there are four major types of drilling fluids including water-based, oil-based, synthetic-based, and pneumatic drilling fluids. Synthetic-based drilling fluid (SBDF) and oil-based drilling fluid (OBDF) are preferred due to their good shale stabilization ability, high lubricating properties, and minimal effect of temperature on them [3,4]. However, their use continues to dwindle due to calls from the government and other agencies on their environmental toxicity as well as their non-conformity with certain aspects of the sustainable development goals (SDGs) [5]. This makes WBDF particularly attractive regardless of its limitations compared to OBDF and SBDF.

For these reasons, nanoparticles and surfactants have been studied to enhance the performance of WBDFs to attain comparable levels as OBDFs and SBDFs. Currently, nanoparticles have shown to be feasible in improving the quality of WBDFs [6]. In drilling fluids, their ability to affect rheology, fluid loss, thermal stability, wellbore stability, shale stability, surface tension, and lubrication has been studied [4]. In terms of its application in drilling fluid rheology and fluid loss, their strong inter-particle relation causes them to affect the physical properties of the fluid making them potentially good for increasing viscosity and decreasing fluid loss [7]. Some researchers [8,9] have observed positive results in rheology when using them. Nanoparticles possess the ability to seal the movement of water between the pore spaces in the formation by plugging into those pores making them effective in fluid loss control [10,11]. Nanoparticles have also been observed to result in stable rheology of fluids subjected to high temperatures; this ensures thermal stability and thus prevents loss of circulation, barite sag, and differential sticking [12]. Similarly, their use has expanded to surface tension reduction in fluids [13,14].

Surfactants have also been studied to improve drilling performance as they are good viscosifying agents in drilling fluids [15].

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The viscosity of the fluid is said to be generated by the self-assembly of the surfactant molecules forming long molecular chains in aqueous solutions [16]. Gemini surfactants belong to a group of surfactants that possess two conventional surfactant molecules linked by a spacer [17]. This feature makes Gemini surfactants behave essentially like two surfactants in one making their performance much better than the corresponding single chained surfactants with equal length. They possess the ability to aggregate at a much lower concentration, that is, critical micelle concentration (CMC), and thus are more active at fluid surfaces as compared to conventional surfactants. The lower CMC means they are more effective in surface tension reduction and better in increasing the viscosity of drilling fluid [18]. One conventional surfactant and one Gemini surfactant were used in this study. Surfactants are characterized as having a polar/hydrophilic head group and a non-polar/hydrophobic tail or chain. In the case of Gemini surfactants, there are two surfactant molecules joined together by a spacer which is a hydrocarbon chain and can be of varying lengths.

While some studies have examined certain surfactants and nanoparticles individually to affect viscosity, surface tension, filtration loss, and thermal stability, few have studied the combination of both, and none reported has specifically applied Gemini surfactants nor its combination with nanoparticles to study their combined effects on the aforementioned WBDF properties. The objective of this study, therefore, is to investigate the synergistic effects of silica nanoparticles and Gemini surfactants on the properties of WBDF.

2. Material and methods

2.1. Materials

Table 1 shows the list of chemicals used in this study while Table 2 shows the detailed description of surfactants.

2.2. Preparation of surfactant solutions and surfactant-silica nanofluids

Gemini surfactants were dispersed in 100 mL of de-ionized water and stirred at 600 rpm for 45 min to gain a homogeneous solution before being added to the base drilling fluid. For the preparation of surfactant-silica nanofluid, silica nanofluid was first prepared by dispersing the nanoparticles in 50 mL of de-ionized water at room temperature before adding to the pre-prepared surfactant solutions. The homogeneity of the solutions was achieved via 30 min of ultra-sonication dispersion (Ultrasonic bath FB15051, Fisher brand). 1 wt% of surfactant solutions and surfactant-silica nanofluids were added to the base drilling fluid before each experimentation.

Table 1
List of chemicals used in the study including their source and purpose.

Material	Source of material	Purpose in the experiment
SF3	Zhejiang Runhe Chemical New Material Co., Ltd. (Hangzhou, China)	Surfactant
SF5	Lion Specialty Chemicals Co., Ltd. (Tokyo, Japan)	Surfactant
SiO_2 nanoparticles (10–15 nm average size)	Tecnan (Tokyo, Japan)	Nanoparticle
Bentonite natural clay, sodium hydroxide pellets, sodium carbonate, and barite	Irama Canggih Sdn Bhd (Perak, Malaysia)	Drilling fluid formulation
High viscosity (HV) and Low viscosity (LV) grade Carboxymethyl Cellulose (CMC)	Henan Botai Chemical Building Materials Company Ltd. (Zhengzhou, China)	Drilling fluid formulation

Table 2
Chemical names of surfactants used and a description of the type of surfactant.

Surfactant Codename	Surfactant chemical name	Description
SF1	3-(2-methoxyethoxy)propyl-methyl-bis(trimethylsilyloxy)silane	Non-ionic silane-based surfactant
SF2	Trimethylene-1,3-bis (hexadecyl dimethyl-ammonium bromide)	Cationic Gemini surfactant

2.3. Water-based drilling fluid preparation

The WBDF was prepared by first adding 0.35 g of soda ash for calcium ion contamination treatment and the same amount of caustic soda to increase and maintain the pH of the drilling fluid at an acceptable range of 8.5 to 10 in 350 mL of distilled water. After five minutes, 14 g, 2.45 g, and 1.05 g of bentonite, LV CMC, and HV CMC were added to the mixture respectively to increase its viscosity and filtration properties. This mixture was also stirred for 5 min. Finally, 112 g of barite was added as a weighting agent and the drilling fluid was stirred for 30 mins to achieve a 10^{+} lb/gal homogenous sample.

2.4. FTIR Characterization

The FTIR spectroscopy of the samples was measured by the Nicolet iS5 FTIR spectrometer (Thermo Scientific, Malaysia). This measures the frequency of light in the infrared region absorbed by the bonds of different chemical elements to determine its molecular composition and structure.

2.5. Viscosity measurement

Viscosity measurement was carried out using Brookfield's DV-III Ultra programmable rheometer. Viscosity measurements were taken for rotation speeds in the range of 20 rpm to 240 rpm (shear rate from 24.5 to 293.5 1/s) using a vane spindle (V-73). Thereafter, the experimental data were fitted to the Herschel-Bulkley model and power-law of viscosity model described in Eq. 1 and 2 to analyze the fluid's properties.

Herschel-Bulkley model:

$$\tau = \tau_y + k\gamma^n \quad (1)$$

Power-law rheology model:

$$\tau = k\gamma^n \quad (2)$$

where τ = shear viscosity; γ = shear rate; τ_y = Yield-stress; k = consistency index; and n = power-law index

The thermal stability was measured by calculating the average change in viscosity as the temperature of the drilling fluid was increased from 25 °C to 150 °C (25 °C, 50 °C, 100 °C, and 150 °C); the lowest change meaning the fluid viscosity experienced the least effect of temperature increase on its viscosity, thus higher thermal stability.

2.5.1. Drilling fluid filtration loss measurement

This fluid property was measured after the WBDF was mixed with each surfactant solution and surfactant-silica nanofluid. The drilling fluid was filtered using an LPLT filter press. An applied pressure of 100 psi was exerted on the fluid thereby forcing the water to be drained from it. This was done at room temperature (26 °C) for thirty minutes before the fluid loss was measured.

2.6. Surface tension measurement

The surface tension was measured for each fluid sample that was prepared was performed using a Du Noüy ring method (SEO-DST30M tensiometer). It consists of a platinum-iridium ring that measures the amount of force required for the ring to be lifted from the surface of the fluid after it has been immersed in it. This force is the surface tension of the fluid. All measurements were done at room temperature (26 °C).

3. Results and discussion

3.1. FTIR spectroscopy

SiO_2 was confirmed in the solution containing Gemini surfactant by the FTIR spectra. Fig. 1 shows the FTIR spectra of drilling fluid with surfactant and with surfactant-silica nanofluid. The peak at 1058.98 cm^{-1} belongs to the stretching and bending vibrations of Si—O—Si which indicates the presence of silica nanoparticles in the WBDF indicating that it has reacted and been incorporated into the drilling fluid; the peak region for SiO_2 usually lies between 1000 and 1250 cm^{-1} [19]. The strong intensity band at 3328.08 cm^{-1} and medium band at 1633.85 cm^{-1} which is close to the deformation band of molecular water belongs to the hydroxyl (–OH) group indicating the presence of water in the fluid [20]. In both spectrums, the weak 981.35 cm^{-1} absorption bands are assigned to the symmetric C–H stretching vibration associated with the backbone of the surfactant [21].

3.2. Effect of surfactant on fluid viscosity

Fig. 2 illustrates the viscosity of the WBDF after the addition of two different types of surfactants. Based on the fitted Power-law and Herschel-Bulkley equations, the corresponding parameters for each equation are presented in Table 3.

As illustrated in Fig. 2, the fluid viscosity increased with the addition of surfactant, with Gemini surfactant expressing the higher increment showing up to 201% increase in yield stress and a 639% increase in the consistency index of the drilling fluid. The shear rate to shear viscosity relationship showed that the viscosity of the WBDF decreased when the shear rate increased indicating a thixotropic fluid. This flow behavior is indicated by the power-law index (n) and it describes a shear-thinning fluid ($n < 1$), as viscosity is dependent on the shear rate; with viscosity decreasing with increasing shear rate [20]. As the result indicated, Yield-stress (τ_y) increased when surfactants (SF1 and SF2) were added; SF2 (Gemini surfactant) showed the highest increment compared to the conventional surfactant. Drilling fluids are expected to suspend drill cuttings and other weighting agents under static conditions; this is made possible by the presence of a yield-stress and this parameter indicates the drilling fluid's suitability for hole cleaning and preventing barite sag [22,23]. Similarly, the power-law model shows an increase in $\boldsymbol{K}$ when surfactants were added to the drilling fluid. When surfactants are added to water, they aggregate to form micelles at a critical micelle concentration due to the water molecules bonding among themselves with minimal interaction with the hydrophobic part of the surfactant. This unique behavior and the arrangement of the surfactants into micelles is responsible for the development of viscosity in the fluid which is more in Gemini surfactants. Due to their structure, Gemini surfactants (containing two surfactant moieties) form more micelles compared to conventional surfactants and this would lead to a higher increase in viscosity. The formation of micelles improves the fluid viscosity

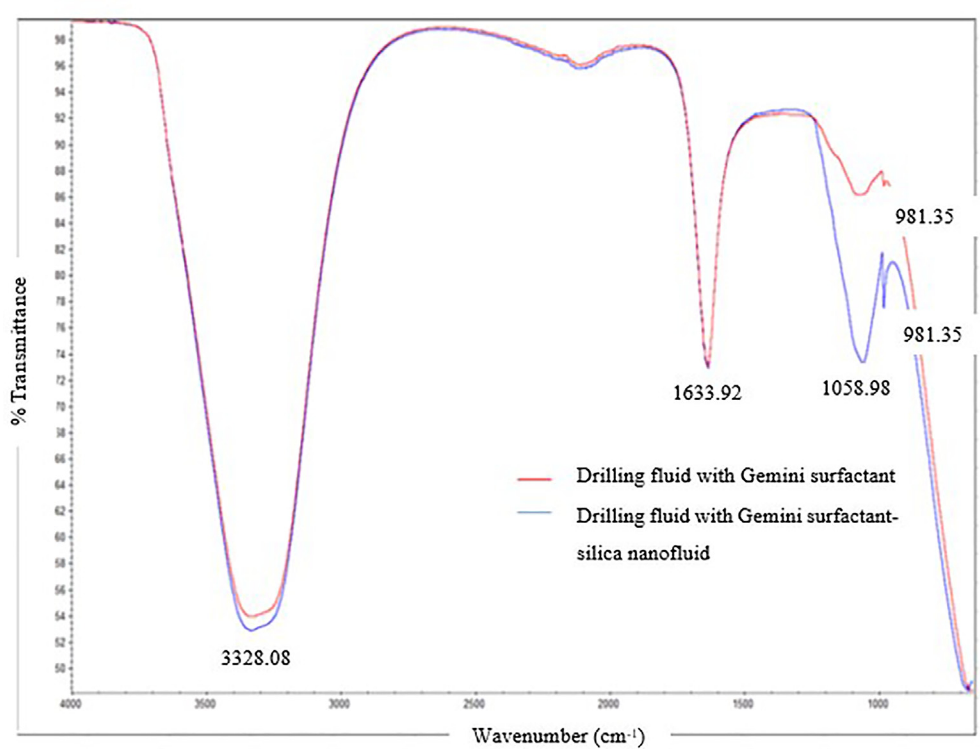

Fig. 1. FTIR spectra of the drilling fluid with and without silica nanofluid.

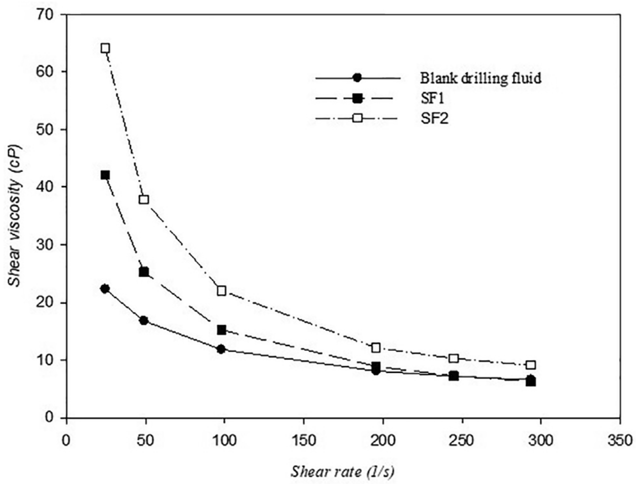

Fig. 2. Viscosity of the drilling fluid containing different surfactants.

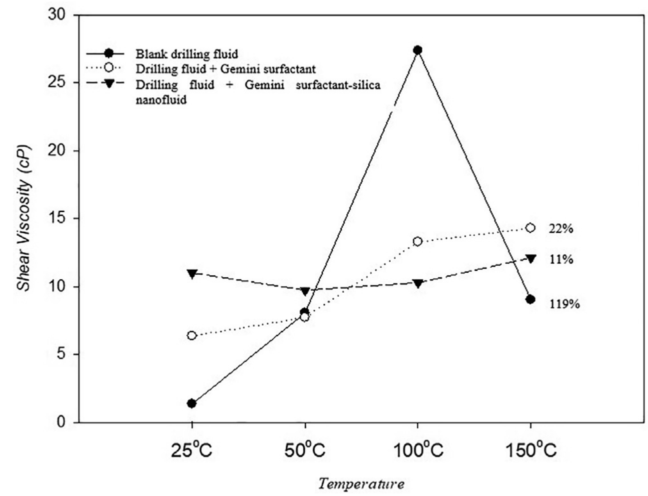

Fig. 3. Change in drilling fluid viscosity with increasing temperature.

because of the entanglement of the long and flexible micelle aggregates to form a transient network in the solution [16,24].

3.3. Effect of Gemini surfactant – silica nanofluid on the thermal stability of WBDF

A comparison of the thermal stability of three formulations of WBDF was carried out by heating at a constant shear rate (293.52/s) for all three samples. As seen in Fig. 3 and Table 4, the blank WBDF and the one with only Gemini surfactant added showed higher changes to its viscosity in comparison to the WBDF containing Gemini surfactant–silica nanofluid. Specifically, the calculated average change in viscosity from 25℃ to 150℃ was recorded at 119%, 22%, and 11% for blank drilling fluid, drilling fluid with Gemini surfactant, and drilling fluid with Gemini surfactant-silica nanofluid respectively. The improvement in thermal stability could be a result of the Brownian motion of nanoparticles, formation of packed structures in nanofluids, and clustering of the nanoparticles when in solution. It helps in forming a coat around the drilling fluid particles thereby protecting them from the effect of high temperature, thus preventing degradation [25]. Drilling fluids tend to degrade at high temperatures, thereby damaging the fluid and leading to several issues during drilling operation including loss of circulation, stuck pipe, and torque & drag.

3.4. Drilling fluid filtration studies

The drilling fluid filtration loss (fluid loss), as well as cake thickness measurements, were done after thirty minutes using an LPLT filter press and digital Vernier caliper respectively. Combinations of Gemini surfactant with silica nanofluid at different concentrations were applied to observe their effects on the WBDFs' fluid loss as presented in Table 5. The experimental results showed that when nanoparticles were added to the system there was a decrease in the fluid loss and a further decrease when the concentration of the nanoparticle was increased. No changes to the filter cake thickness were observed. Nanoparticles tend to plug into the pore spaces in-between solid particles thereby making it compact and retaining the water content of the fluid [26], thus the decrease in fluid loss.

Table 4
Percentage change in viscosity with increasing temperature.

	Blank drilling fluid (%)	Drilling fluid + Gemini solution (%)	Drilling fluid + Gemini surfactant – silica nanofluid (%)
25-50℃	83	18	13
50-100℃	71	42	5
100-150℃	204	7	15
Avg.	119	22	11

3.5. Surface tension analysis

Fig. 4 shows the surface tension for four samples of drilling fluid. There was a 5% reduction in the surface tension of the WBDF when a conventional surfactant (SF1) was added but using Gemini surfactant further reduces the surface tension by 8%. More importantly, the reduction in surface tension became even significant in the presence of silica nanofluid (17% reduction). Gemini surfactants are characterized as having two surfactant moieties linked by a spacer. This makes them more effective surface tension reducers relative to the corresponding conventional surfactant of equal chain length because they possess much lower critical micelle concentration values [27]. When combined with silica nanofluid in the drilling fluid it showed the most reduction in surface tension. This occurs due to the behavioral mechanism of nanoparticle absorption with surfactants; as the surfactants begin to absorb the nanoparticles, they are effectively pushed to the fluid's surface, and the presence of more surfactant molecules at the surface

Table 3
Viscosity parameters for the fitted Herschel-Bulkley and power-law model.

Sample	Herschel-Bulkley model		Power-law model		
	τ_y (cP)	R^2	K (Pa.S^n)	|n|	R^2
Blank drilling fluid	41.79	0.9885	114.15	0.5	0.9975
SF1	82.31	0.9511	493.16	0.764	0.9994
SF2	125.92	0.9453	832.28	0.797	0.9995

Table 5
Fluid loss and filter cake thickness of the drilling fluid containing different formulations of Gemini surfactant-silica nanofluid.

Sample	Fluid loss (mL)	% decrease	Filter cake (mm)	% decrease
Blank drilling fluid	8.5	–	2	0
Sample 4 (400 ppm Gemini surfactant)	8.5	0	2	0
Sample 5 (400 ppm Gemini surfactant + 100 ppm silica nanofluid)	8	5.88	2	0
Sample 6 (400 ppm Gemini surfactant + 300 ppm silica nanofluid)	7.5	11.76	2	0

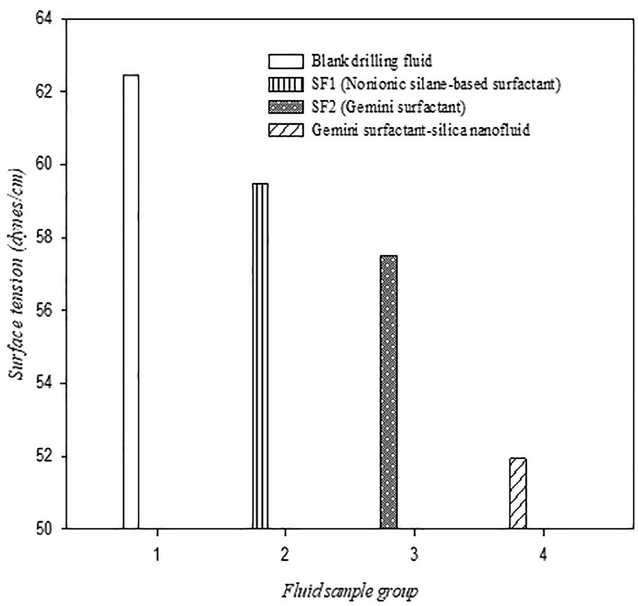

Fig. 4. Surface tension of drilling fluid containing different surfactants and with silica nanofluid.

causes a better surface tension reduction [28]. Lower surface tension in drilling fluid helps in reducing drilling problems like stuck pipe and enhancing oil recovery.

4. Conclusion

In this study, Gemini surfactant-silica nanofluids were formulated and then used as an additive in the WBDF. The fluid samples were characterized by FTIR analysis to confirm the formulation of the fluid samples. The effects of this sample in WBDF were studied. It was observed that the Gemini surfactant increased the viscosity of the WBDF as shown from the increase in yield-stress and consistency index by 201% and 639% respectively. When combined with the silica nanofluid it helps to improve the thermal stability of the drilling fluid with an average of 11% change in viscosity. Furthermore, the silica nanofluid improves the filtration loss property by inducing a reduction in the fluid loss by 12%. The Gemini surfactant-silica nanofluid induced a higher reduction (17%) in the surface tension of the drilling fluid.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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